Studying the High-rate Deformation of Soft Materials via Laserinduced Membrane Expansion

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Supporting Information

I. LIME EXPERIMENTS

Certain instruments and materials are identified in this paper to adequately specify the experimental details. Such identification does not imply recommendation by the National Institute of Standards and Technology; nor does it imply that the materials are necessarily the best available for the purpose.

A. Materials

Polydimethylsiloxane (PDMS, Sylgard 184) was purchased from Dow Chemicals. 20 cm x 40 cm glass cover slips were purchased from Corning Glass.

B. Ablation Target Preparation

Ablation targets were prepared by first sputter coating 30 nm of gold on a glass cover slip. Next, PDMS was mixed, degassed for 30 m, and subsequently deposited via spin-coating between 99 to 315 rad/s for 90 seconds to form an elastomeric layer (10 to 60 μ m). The deposited films were further degassed for another 30 m and finally thermally cured at 70 °C for 2 h.

C. Laser-induced membrane expansion (LIME) test

A pulsed diode-pumped solid-state infrared laser (Flare NX, $\lambda = 1030$ nm, pulse length = 1.5 ns, Coherent Inc) was focused on the gold layer of an ablation target. An ultra-fast camera (Specialized Imaging Ltd, SIMD12) was used to capture the membrane expansion process with 20 ns exposure time per frame and an interframe time of 140 ns (10, 22, 35, and 45 µm films) or 340 ns (60 µm films). The camera has a maximum of 12 frames and error of the camera is ± 3 ns. Synchronization of the ablation event and image acquisition was achieved via digital triggers modulated using a digital waveform generator (NI-9402, National Instruments). Each image collected was subjected to background subtraction and thresholding to eliminate noise using ImageJ. The position of the expanding membrane was measured using Tracker Video Analysis and Modeling Tool. The radius

reported was that of a hemispherical cap, with each image captured being taken as a cross-section of that cap. The radius itself was taken as the center point of the membrane to the furthermost point on the cross-section in the vertical direction, measured along the center axis.

II. BUCKINGHAM II ANALYSIS FOR MEMBRANE INFLATION

A. Bubble Expansion in the Absence of a Membrane

The theoretical maximum rate of expansion of a bubble due to laser ablation is analogous to the expansion of a shock wave in a nuclear explosion. In a nuclear explosion, there is an instantaneous release of energy U in a small region of space. This produces a spherical shock wave, with the pressure inside the shock wave several thousands of times greater than the initial air pressure, which can be neglected.

Using Buckingham Π analysis, we can relate the radius (R) of this shock wave to time (t). The relevant parameters are U, R, air density (ρ_m) and t. Thus,

$$R = R(U, \rho_{\rm m}, t) \tag{S.1}$$

There are n = 4 physical variables and j = 3 dimensions, and therefore 3 repeating variables. Thus, there are $n - j = 4 - 3 = 1 \prod_i$ group. Table I summarizes the variables and the corresponding dimensions.

Variable	Description	Dimensions
R	radius of blast	[L]
U	blast energy	$[ML^2/T^2]$
$ ho_{ m m}$	air density	$[M/L^3]$
t	time	[T]

TABLE I. Variables and corresponding dimensions for bubble expansion in an air medium.

The first and only Π_i group is defined as,

$$\Pi_1 = R U^{\alpha} \rho_{\rm m}^{\beta} t^{\gamma}$$

= [L][ML²/T²]^{\alpha}[M/L³]^{\beta}[T]^{\gamma} (S.2)

Note that since j = 3, there are 3 repeating variables. We are interested in solving for R, we do not want to have R as one of the repeating variables. From Eq.(S.2), we can develop a system of equations to determine the power law coefficients.

$$[L][ML2/T2]\alpha[M/L3]\beta[T]\gamma = [L]0[M]0[T]0$$
(S.3)

$$\begin{split} [L] &: 1 + 2\alpha - 3\beta = 0 \implies \alpha = -1/5 \\ [M] &: \alpha + \beta = 0 \implies \beta = 1/5 \\ [T] &: -2\alpha + \gamma = 0 \implies \gamma = -2/5 \end{split} \tag{S.4}$$

Therefore, Eq.(S.2) becomes,

$$\Pi_1 = RU^{-1/5} \rho_{\rm m}^{1/5} t^{-2/5} \tag{S.5}$$

Solving for R, we obtain the classic relationship for Taylor's blast,

$$R = c_0 \left(\frac{U}{\rho_{\rm m}}\right)^{1/5} t^{2/5}$$
 (S.6)

B. Bubble Expansion within a Semi-infinite Elastic Medium

This section discuss the relevant parameters for seededlaser induced cavitation experiments Here, the radius Rof this bubble is related to the previous parameters of U, $\rho_{\rm p}$, t as well as the tension of the membrane ($\gamma_{\rm el}$), which has units of force/length and is related to the strain energy density ($W_{\rm el}$) and thickness of the membrane (h). Since the medium is semi-finite, h is not a relevant parameter. Thus,

$$R = R(U, \gamma_{\rm el}, \rho_{\rm p}, t) \tag{S.7}$$

There are n = 5 physical variables and j = 3 dimensions, and therefore 3 repeating variables. Thus, there are $n - j = 5 - 3 = 2\Pi_i$ groups. Table II summarizes the variables and the corresponding dimensions.

Variable	Description	Dimensions
R	radius of cavitation	[L]
U	blast energy	$[ML^2/T^2]$
γ_{el}	membrane tension	$[M/T^2]$
$ ho_p$	membrane density	$[M/L^3]$
t	time	[T]

TABLE II. Variables and corresponding dimensions for bubble expansion within a semi-infinite elastic medium.

For Π_1 , we choose U, ρ_p, t as the 3 repeating variables and R as the dependent variable. The result is simply Eq.(S.5),

$$\Pi_1 = R \left(\frac{\rho_{\rm p}}{U}\right)^{1/5} t^{-2/5} \tag{S.8}$$

For Π_2 , we choose U, ρ_p, t as the 3 repeating variables and γ_{el} as the dependent variable.

$$\Pi_2 = \gamma_{\rm el} U^{\alpha} \rho_{\rm p}^{\beta} t^{\gamma}$$
$$= \gamma_{\rm el} U^{-3/5} \rho_{\rm p}^{-2/5} t^{4/5}$$
(S.9)

Buckingham Π theorem tells us that $\Pi_1 = \Phi(\Pi_2)$. Experimentally, we know that $R \sim t^{\alpha}$ at short times, and $R \sim t^{-\beta}$ at long times. Therefore, $\Phi(\Pi_2) = c_1$ when $R < R_c$, and $\Phi(\Pi_2) = We^*$ when $R > R_c$. Here, We^* is similar to a Weber number, which compares the importance between inertia and interfacial tension, except here we are dealing with interfacial tension due to elasticity of the material. One possible solution is,

$$c_1 \Pi_1 + c_2 \Pi_2 = c_3 \tag{S.10}$$

Substituting the above expressions in Eq.(S.10), we obtain,

$$R = c_1 \left(\frac{U}{\rho_p}\right)^{1/5} t^{2/5} - c_2 \left(\frac{\gamma_{el}}{U^{3/5} \rho_p^{2/5}}\right) t^{6/5} \qquad (S.11)$$

Eq.(S.11) suggests that the inertia dominates the cavitation process at short times or when $R < R_c$, and membrane tension ($\gamma_{\rm el}$) dominates the expansion process at long times or when $R > R_c$. In a laser ablation experiment, the laser power is a fixed quantity, therefore, c_1 , c_2 , U and $\rho_{\rm p}$ are constants. Hence, we should be able to determine the density of the medium at short times and elasticity of the medium by relating R to t at long times.



FIG. S1. Radius of expanding bubble (R) as a function time (t) for laser-induced cavitation (LIC) of air and elastomer. At early times, the growth of both kinds of bubble is dominated by inertia of the medium. Only at late times is the bubble growth affected by the elasticity of the elastomer.

C. Bubble Expansion of a Thin Membrane

Using the insights gained from the previous section, we can develop scaling relationships for the rapid expansion of a thin elastomeric membrane that was initially flat. The radius of the expanding membrane is related to the following variables,

$$R = R(h, U, \gamma_{\rm el}, \rho_{\rm m}, \rho_{\rm p}, t) \tag{S.12}$$

There are n = 7 physical variables and j = 3 dimensions, and therefore 3 repeating variables. Thus, there are $n - j = 7 - 3 = 4\Pi_i$ groups as summarized in Table III.

For Π_1 , we choose again U, ρ_p , t as the 3 repeating variables and R as the dependent variable, which is Eq.(S.5),

$$\Pi_1 = R \left(\frac{\rho_{\rm p}}{U}\right)^{1/5} t^{-2/5} \tag{S.13}$$

Variable	Description	Dimensions
R	radius of expansion	[L]
h	membrane thickness	[L]
U	blast energy	$[ML^2/T^2]$
$\gamma_{ m el}$	membrane tension	$[M/T^2]$
$ ho_{ m m}$	surrounding medium density	$[M/L^3]$
$ ho_{ m p}$	membrane density	$[M/L^3]$
t	time	[T]

TABLE III. Variables and corresponding dimensions for bubble expansion of a thin membrane.

For Π_2 , we again choose U, ρ_p , t as the 3 repeating variables and γ_{el} as the dependent variable, which is Eq.(S.9),

$$\Pi_2 = \gamma_{\rm el} U^{-3/5} \rho_{\rm p}^{-2/5} t^{4/5} \tag{S.14}$$

For Π_3 , we choose U, ρ_p, t as the 3 repeating variables and h as the dependent variable,

$$\Pi_3 = h \left(\frac{\rho_{\rm p}}{U}\right)^{1/5} t^{-2/5} \tag{S.15}$$

For Π_4 , we choose $U, \rho_{\rm p}, t$ as the 3 repeating variables and $\rho_{\rm m}$ as the dependent variable,

$$\Pi_4 = \left(\frac{\rho_{\rm m}}{\rho_{\rm p}}\right)^n \tag{S.16}$$

since we don't know exact value of the scaling exponent n. Based on the derivation for bubble expansion of a semi-infinite medium, a possible solution is,

$$c_1\Pi_1 + c_2\Pi_2 + c_3\Pi_3 = c_4\Pi_4 \tag{S.17}$$

Substituting all the above expressions into Eq.(S.17), we obtain,

$$c_{1}R\left(\frac{\rho_{\rm p}}{U}\right)^{1/5}t^{-2/5} + c_{2}\frac{\gamma_{\rm el}}{U^{3/5}\rho_{\rm p}^{2/5}}t^{4/5} + c_{3}h\left(\frac{\rho_{\rm p}}{U}\right)^{1/5}t^{-2/5} = c_{4}\left(\frac{\rho_{\rm m}}{\rho_{\rm p}}\right)^{n}$$
(S.18)

Note that although this expression appears to be significantly different than Eq.(S.11), they share similar constraints including resistance due to inertia and resistance due to elasticity of the material. The expressions deviate with the additional Π_3 and Π_4 terms, which are related to correction for the finite size of the membrane and mechanical impedance mismatch of membrane and surrounding medium.

We can estimate the $\gamma_{\rm el}$ by assuming a specific hyperelastic model. From Laprade and coworkers, membrane tension is related to the stretch ratio ($\lambda = R/R_{\rm o}$),

$$\gamma_{\rm el} = \frac{h}{\lambda} \frac{\partial W_{\rm el}}{\partial \lambda} \tag{S.19}$$

The exact form of $W_{\rm el}$, such as the Neohookean or Mooney-Rivlin model, depends on the specific material. We can estimate the $\gamma_{\rm el}$ by assuming a specific hyperelastic model. The biaxial expansion of a membrane according to the Neohookean model is defined as,

$$\gamma_{el} = \frac{h}{\lambda} \mu (\lambda^2 - \lambda^{-4})$$

$$\approx \frac{Rh}{R_o} \mu$$
(S.20)

where $\lambda = R/R_{o}$, and μ is the shear modulus of the membrane. Substituting Eq.(S.20) into Eq.(S.18), we obtain,

$$R = \frac{c_4 \left(\frac{\rho_{\rm m}}{\rho_{\rm p}}\right)^n t^{2/5} - c_3 h \left(\frac{\rho_{\rm p}}{U}\right)^{1/5}}{c_1 \left(\frac{\rho_{\rm p}}{U}\right)^{1/5} + c_2 U^{-3/5} \rho_{\rm p}^{-2/5} \left(\frac{h}{R_{\rm o}}\right) \mu t^{6/5}}$$
$$= \frac{c_4 \left(\frac{U}{\rho_{\rm p}}\right)^{1/5} \left(\frac{\rho_{\rm m}}{\rho_{\rm p}}\right)^n t^{2/5} - c_3 h}{1 + c_2 U^{-2/5} \rho_{\rm p}^{-3/5} \left(\frac{h}{R_{\rm o}}\right) \mu t^{6/5}}$$
$$= \frac{C_4 t^{2/5} - C_3 h}{1 + C_2 \left(\frac{h}{R_{\rm o}}\right) \mu t^{6/5}}$$
(S.21)